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Optical properties of GaN with Ga and N polarity

M. A. Reshchikov, D. Huang, F. Yun, P. Visconti,[a] T. King, and H. Morkoç
Virginia Commonwealth University, Richmond, VA 23284, U.S.A.
[a] Also with Instituto per lo Studio di Nuovi Materiali per l'Elettronica, CNR, Via Arnesano, Lecce, Italy

J. Jasinski [b] and Z. Liliental-Weber
Lawrence Berkeley National Laboratory, Berkeley, CA 94720, U.S.A.
[a] Also with the Institute of Experimental Physics, Warsaw University, Warsaw, Poland

ABSTRACT

We compared photoluminescence (PL) and cross-sectional transmission electron microscopy (TEM) characteristics of GaN samples with Ga and N polarities grown by molecular beam epitaxy (MBE) on sapphire substrates. Ga-polar films grown at low temperature typically have very smooth surfaces, which are extremely difficult to etch with acids or bases. In contrast, the N-polar films have rougher surfaces and can be easily etched in hot H_3PO_4 or KOH. The quality of the X-ray diffraction spectra is also much better in case of Ga-polar films. Surprisingly, PL efficiency is always much higher in the N-polar GaN, yet the features and shape of the PL spectra are comparable for both polarities. We concluded that, despite the excellent quality of the surface, MBE-grown Ga-polar GaN layers contain higher concentration of nonradiative defects. From the analyses of cross-sectional TEM investigations, we have found that Ga-polar films have high density of threading dislocations (5×10^9 cm^{-2}) and low density of inversion domains (1×10^7 cm^{-2}). For N-polar GaN the situation is the reverse: the density of dislocations and inversion domains are 5×10^8 and $\sim 1 \times 10^{11}$ cm^{-2}, respectively. One of the important conclusions derived from the combined PL and TEM study is that inversion domains do not seem to affect the radiative efficiency very adversly, whereas dislocations reduce it significantly.

INTRODUCTION

Wurtzite GaN is a polar crystal. When it is grown on *c*-plane of sapphire substrates, the crystal direction [0001] of the resulting GaN film can be either parallel or antiparallel to the growth direction, leading to epilayers with Ga polarity (the (0001) plane is Ga terminated) or N polarity (the (000-1) plane is N terminated). In MBE growth, Ga-polar films are usually obtained on AlN buffer layers and N-polar films - on GaN buffers [1]. Previous investigations showed that the films with different polarities have very different structural and electronic properties [2]. Ga-polar films have relatively smooth and stable surface, which is very difficult to etch. The PL properties of Ga-polar films grown by MOCVD also seem better than N-polar films [3,4]. It is often believed that the overall quality of the Ga-polar films is much better than N-polar films. However, N-polar films may be important for some device applications [5,6]. In this paper, we analyze the structural and optical characteristics of Ga- and N-polar GaN films grown by MBE.

EXPERIMENTAL DETAILS

Undoped GaN layers with thickness from 1 to 3 μm were grown on *c*-plane of sapphire by MBE using radio frequency activated nitrogen. The layers with Ga polarity were grown on AlN buffer, N-polar films were grown on GaN buffer. In some samples, a 10 period GaN/AlN superlattice of total thickness of ~30 nm was inserted in the epilayer to improve the crystalline quality [7]. The quality of the layer structure was examined by high-resolution X-ray diffraction

(XRD) and atomic force microscopy (AFM) in conventional tapping mode. Etching in H_3PO_4 at 160ºC for the time from 15 sec. to 15 min. was used to reveal pits originating from threading dislocations and to establish etching rate. The polarity of the samples was established from detailed analyses including in-situ RHEED patterns, X-ray data, AFM images before and after etching. Convergent beam electron diffraction (CBED) studies [8,9] were also applied to a few of the samples, which confirmed the polarity assignment based on RHEED and etching experiments [10]. In addition, conventional TEM studies were carried out in order to examine the details of the microstructure of the films. Both CBED and TEM studies were performed using a TOPCON 002B electron microscope operated at 200 keV [11].

For a detailed analysis of optical properties, we have chosen two representative samples with Ga polarity (sample G) and N polarity (sample N). PL was excited with a He-Cd laser and analyzed with a SPEX grating monochromator and Hamamatsu photomultiplier tube. Excitation density was 0.3 W/cm^2. The sample temperature during the measurements was varied between 15 and 300 K using a closed cycle He cryostat.

RESULTS AND DISCUSSION

Figure 1 presents typical AFM images for the as-grown surfaces of the Ga- and N-polar GaN films prepared by MBE. N-polar layers contain tall columns with a very flat surface separated by deep troughs. The surface of the Ga-polar film is much flatter with visible defects that apparently convert into hexagonal-shaped pits after etching in hot H_3PO_4. From the image shown in Fig. 1(a), the defect density in the sample G was estimated to be (2-6)$\times 10^9$ cm^{-2}.

(*a*) (*b*)

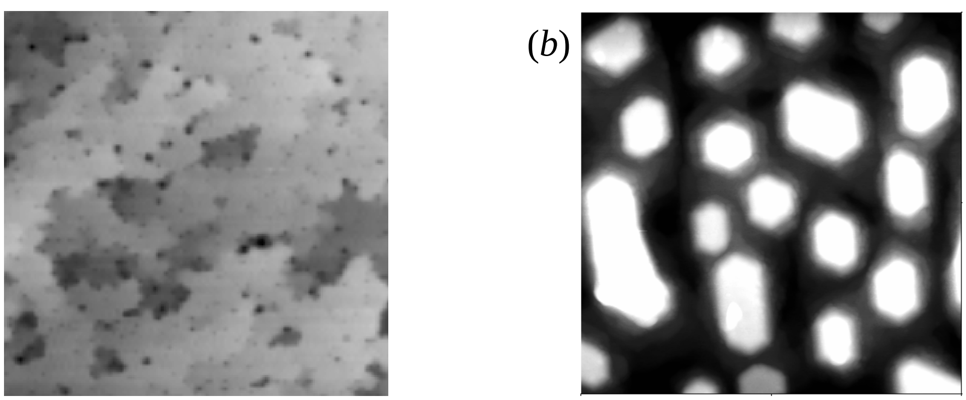

Figure 1. AFM images from (a) the Ga-polar sample G and (b) the N-polar sample N. The image sizes are both 2 × 2 µm. The vertical scales are 15 nm (a) and 70 nm (b). The rms roughness is 1.3 and 23 nm for the images (a) and (b), respectively.

The dislocation density was measured by TEM. Figure 2 presents cross-sectional bright-filed, multi-beam TEM images from the same samples. In the Ga-polar film (Fig. 2(a)), the overall dislocation density is ~1×10^{11}cm^{-2} near the GaN/substrate interface. This density reduces to less than 5×10^9cm^{-2} near the film surface, which is ~1 µm away from the interface. The pit density revealed by chemical etching followed by AFM imaging is consistent with the dislocation density near the film surface for the Ga-polar sample. The TEM images, taken under specific two-beam diffraction conditions, showed that the edge dislocations are the majority and (~95%) of all dislocations in this film. Relatively low density of screw dislocations (<1×10^7cm^{-2}) mixed dislocations (~1×10^8cm^{-2}) were present. The results are very different for the N-polar film,

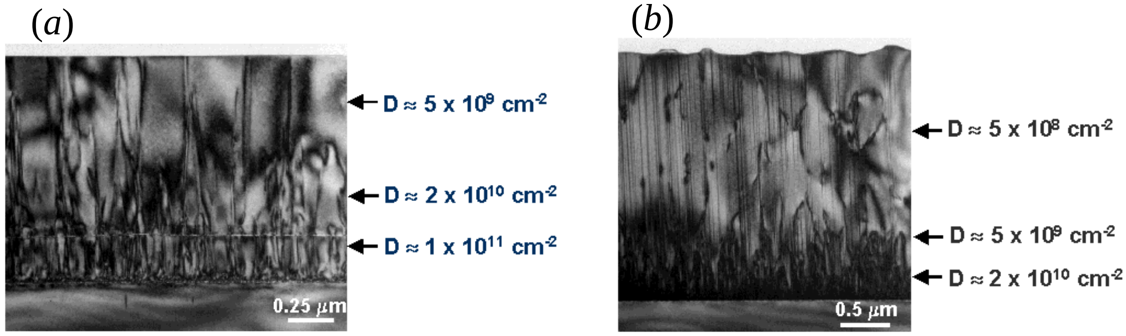

Figure 2. Cross-sectional bright-field TEM images recorded under multi-beam conditions from (a) the Ga-polar sample G and (b) the N-polar sample N.

as shown in Fig. 2(b). In this case, the overall dislocation density is much lower than that observed in the Ga-polar film. The dislocation density is 2×10^{10} cm^{-2} near the GaN/substrate interface and reduces to less than $5\times10^{8}cm^{-2}$ near the film surface. The bright-field TEM images recorded under two-beam conditions showed that all three types of dislocations (edge, screw, and mixed) are present in this N-polar film at comparable densities.

Inversion domains have also been observed in the TEM images of Ga- and N-polar films (Fig. 3). The density of the inversion domains was estimated to be $\sim1\times10^{7}$ and $\sim1\times10^{11}cm^{-2}$ for

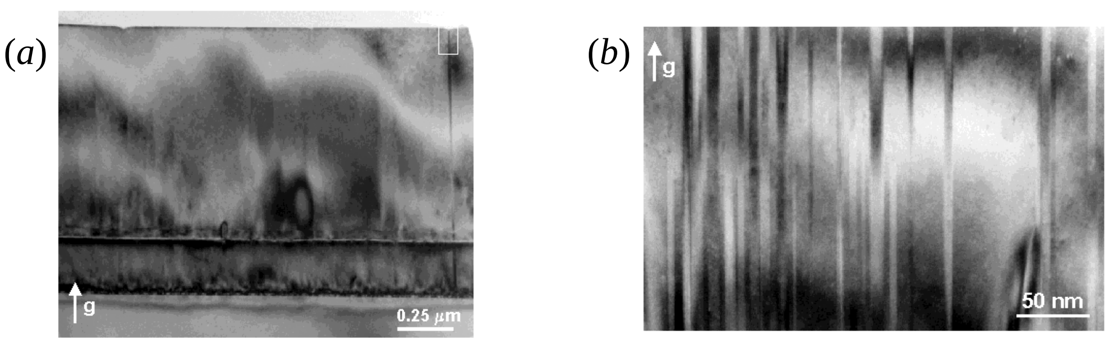

Figure 3. Cross-sectional bright-field TEM images recorded under two-beam conditions (with g-vector parallel to the *c*-axis) from (a) the Ga-polar sample G and (b) the N-polar sample N.

the Ga- and N-polar layers, respectively. All observed inversion domains are wire-like and propagate along the growth direction. They start from the buffer layer and move directly up to the film surface without being interrupted by any intermediate AlN layer. Pits associated with inversion domains were observed in the TEM image of the Ga-polar sample, showing a slightly lower growth rate of the inversion domains as compared to the surrounding Ga-polar matrix [12]. The diameter of the inversion domains ranges from a few to 10 nm. Note that each column and trough observed in the as-grown surface (see Fig. 1(b)) contains many inversion domains as revealed by TEM images. Therefore, no correlation between the surface morphology and the actual inversion domain density [13] can be derived.

The Ga- and N-polar GaN films also showed different XRD characteristics. For the Ga-polar films, narrow (0002) peaks with the full width at half maximum (FWHM) of ~2 arc-minutes were commonly observed. The (10-14) peaks were a few times broader, typically about

5 - 6 arc-minutes. For the majority of N-polar films, the (0002) peak is similar or even broader than (10-14) peak with the FWHM of about 5 - 6 arc-minutes.

Figures 4 (a) and (b) present low- and room-temperature PL spectra from the samples with Ga and N polarity, measured under the same conditions.

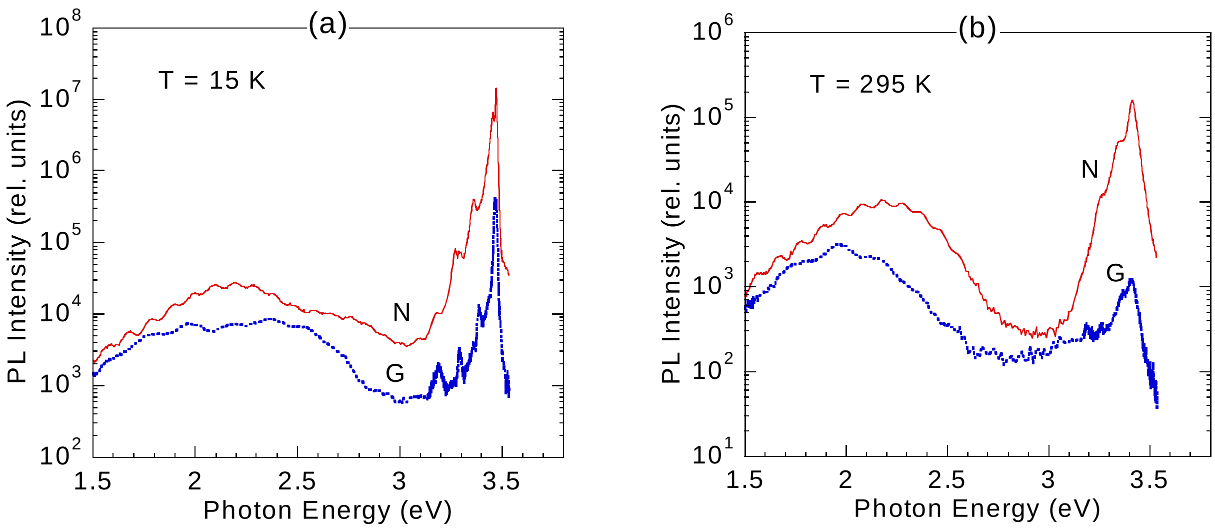

Figure 4. PL spectra of the Ga-polar sample G and the N-polar sample N at low (a) and room (b) temperature. Oscillations below ~ 3.0 eV are due to interference.

For both samples, the donor-bound exciton (DBE) line dominates the spectrum at 15 K. The positions of the DBE lines are 3.463 and 3.471 eV in the Ga- and N-polar films, respectively. The FWHM of these lines is 8-10 meV. The DBE peak quenches with temperature with an activation energy of about 7 meV giving way to the free exciton (FE) line peaking at 6-7 meV higher than the DBE line. The position of the FE line at room temperature is 3.410 and 3.416 eV in the Ga- and N-polar films, respectively. In the sample with N polarity, a peak at 3.455 eV, having a few LO-phonon replicas, is also strong at low temperatures. This peak was previously attributed to the acceptor-bound exciton (ABE) [14]. Recent high spatial resolution PL studies demonstrated that the bright emission at 3.45-3.46 eV originates from the inversion domain boundaries and this peak was attributed to a shallow trap [15]. Our results are consistent with the observations of Ref. 15 since the 3.455 eV peak appears to dominate in the PL spectra of our N-polar samples which have high density of inversion domains. However, to be consistent with the Haynes rule, we believe that excitons bound to some deep-level defects at the inversion domain interface are responsible for the 3.455 eV peak.

While the general features of the spectra (main peaks in terms of their energies and widths, and the relative intensities) are similar for both the Ga- and N-polar films, their absolute PL intensities are very different. For the analyzed samples, the exciton peak from the N-polar film is some 35-100 times stronger than that from the Ga-polar film (Fig. 4). The difference in the dislocation density (5×10^9 cm^{-2} in the Ga-polar film versus 5×10^8 in the N-polar film) may in part be responsible for the much lower PL efficiency in the Ga-polar films. Indeed, for the dislocation densities exceeding 10^7-10^8 cm^{-2}, the radiative emission efficiency in GaN abruptly decreases [16,17].

The TEM investigation indicates that the N-polar film contains a very high density of inversion domains (~ 10^{11} cm^{-2}), about 10^4 times more than the Ga-polar film. Our PL observations, as well as results of Ref. 15, appear to imply that the inversion domains do not reduce the radiative efficiency. Theoretical calculations have shown that there is a low-energy inversion domain boundary in GaN that does not induce electronic states in the band gap and would therefore not affect the PL efficiency [18]. In addition, we assume that the inversion domains in the N-polar films may confine photo-carriers and reduce their non-radiative scattering.

Temperature-related quenching of the excitonic emission was also studied for the samples with Ga and N polarity (Fig. 5).

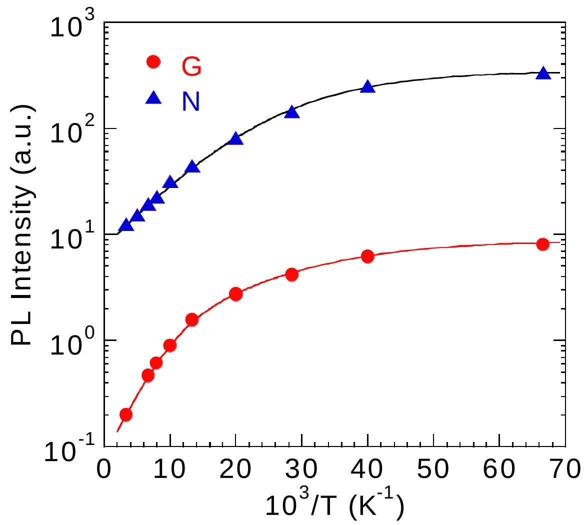

Figure 5. Temperature dependence of the integrated intensity of the excitonic emission from the Ga-polar (G) and N-polar (N) samples.
Solid curves are fit by the expression

$$I^{PL}(T) = \frac{I(0)}{1 + C_1 \exp\left(-\frac{E_1}{kT}\right) + C_2 \exp\left(-\frac{E_2}{kT}\right)}$$

with C_1 = 100 (G) and 35 (N); C_2 = 10 (G and N); E_1 = 26 (G) and 14 (N) meV; E_2 = 7 meV (G and N).

With increasing temperature from 15 to 40-70 K, the integrated intensity of the excitonic emission (in the range of 3.1 – 3.5 eV) quenched with activation energy of about 7 meV for both samples. Quenching can be attributed to detrapping of excitons from the shallow donors. Note that this process did not lead to an increase of the FE emission: the FE emission intensity is temperature independent, even slightly decreases in this temperature range. This means that from different channels of the FE loss (capture by nonradiative centers, capture by shallow donors, radiative recombination, exciton dissociation), the capture of free excitons by nonradiative centers is by far the most efficient channel.

In the temperature range of 70 – 300 K, the excitonic emission intensity in the investigated samples quenches with different activation energies; about 26 meV in the Ga-polar layer and 14 meV in the N-polar layer. Quenching in the Ga-polar layer can be attributed to dissociation of free excitons (the FE activation energy is about 26 meV [19]) followed by trapping of free carriers by nonradiative defects. The mechanism of the quenching with much smaller activation energy in another sample is unclear.

CONCLUSIONS

We have shown that the Ga- and N-polar GaN films grown on sapphire substrates by MBE have very different structural and optical properties. As compared to the Ga-polar films, the N-polar films show rough surfaces, broader (002) XRD peaks, higher density of inversion domains, and are fragile to wet chemical etching. But the N-polar films show a lower density of edge dislocations and a higher PL efficiency. Very low PL efficiency in the Ga-polar layers is related to the high density of threading dislocations. The radiative efficiency in the N-polar layers is up to 100 times higher, in agreement with low density of threading dislocations, which appear to be nonradiative recombination centers. Very high density of inversion domains (~ 10^{11} cm^{-2}) in the N-polar film does not seem to affect the PL efficiency very adversely.

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